

Nodal gap in iron-based superconductor CsFe₂As₂ probed by quasiparticle heat transport

X. C. Hong,¹ X. L. Li,¹ B. Y. Pan,¹ L. P. He,¹ A. F. Wang,² X. G. Luo,² X. H. Chen,² and S. Y. Li^{1,*}

¹*State Key Laboratory of Surface Physics, Department of Physics,*

and Laboratory of Advanced Materials, Fudan University, Shanghai 200433, P. R. China

²*Hefei National Laboratory for Physical Science at Microscale and Department of Physics, University of Science and Technology of China, Hefei, Anhui 230026, China*

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The thermal conductivity of iron-based superconductor CsFe₂As₂ single crystal ($T_c = 1.81$ K) was measured down to 50 mK. A significant residual linear term $\kappa_0/T = 1.27$ mW K⁻² cm⁻¹ is observed in zero magnetic field, which is about 1/10 of the normal-state value in upper critical field H_{c2} . In low magnetic field, κ_0/T increases rapidly with field. The normalized $\kappa_0/T(H)$ curve for our CsFe₂As₂ (with residual resistivity $\rho_0 = 1.80$ $\mu\Omega$ cm) lies between the dirty KFe₂As₂ (with $\rho_0 = 3.32$ $\mu\Omega$ cm) and the clean KFe₂As₂ (with $\rho_0 = 0.21$ $\mu\Omega$ cm), which is consistent with its impurity level. These results strongly suggest nodal superconducting gap in CsFe₂As₂, similar to its sister compound KFe₂As₂.

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I. INTRODUCTION

For the iron-based superconductors,^{1,2} one very important issue is the symmetry and structure of their superconducting gap,³ which is crucial for understanding the mechanism of high-temperature superconductivity.⁴ However, after five-year extensive studies, it is still in a complex situation, mainly due to their multiple electronic bands.^{5,6}

Most of iron-based superconductors have both hole and electron Fermi surfaces, for example Ba_{0.6}K_{0.4}Fe₂As₂.⁶ While many of these superconductors show nodeless superconducting gaps, such as optimally doped BaFe₂As₂,^{6–10} LiFeAs,^{11–14} NaFe_{1-x}Co_xAs,^{15,16} and FeTe_{1-x}Se_x,^{17,18} some of them manifest nodal superconducting gap, such as BaFe₂(As_{1-x}P_x)₂,^{19–21} Ba(Fe_{1-x}Ru_x)₂As₂,²² LiFeP,²³ and LaFePO.^{24,25} So far, it is not conclusive that the nodeless gaps on different Fermi surfaces are s_{\pm} -wave.³ The nodal gap in those iso-valently P- or Ru-substituted compounds could be accidental nodal s -wave on some Fermi surface,²¹ but its origin is still not very clear.²³

More intriguingly, while nodeless superconducting gaps were observed in the extremely electron-deopd A_xFe_{2-y}Se₂ (A = K, Rb, Cs, ...) with only electron pockets,^{26–28} nodal superconducting gap was found in the extremely hole-doped KFe₂As₂ with only hole pockets.^{29,30} It is now under hot debate whether the superconducting gap in KFe₂As₂ is d -wave or accidental nodal s -wave.^{31–33} Thermal conductivity measurements gave compelling evidence for d -wave gap,^{31,32} but recent low-temperature angle-resolved photoemission spectroscopy (ARPES) measurements showed octet-line node structure, suggesting accidental nodal s -wave gap.³³

To clarify this situation in KFe₂As₂, it will be helpful to investigate the superconducting gap structure of its two sister compounds RbFe₂As₂ and CsFe₂As₂, both with $T_c = 2.6$ K from the measurements of

polycrystalline samples.^{34,35} Unexpectedly, recent muon-spin spectroscopy measurements of RbFe₂As₂ polycrystal claimed that the temperature dependence of the superfluid density n_s is best described by a two-gap s -wave model,^{36,37} which is quite different from KFe₂As₂. In this context, more experiments are highly desired, especially on the single crystals of RbFe₂As₂ and CsFe₂As₂.

In this paper, we present the thermal conductivity measurements of CsFe₂As₂ single crystals down to 50 mK. We find clear evidence for superconducting gap nodes from the significant residual linear term κ_0/T in zero field and the field dependence of κ_0/T . Our results suggest common nodal gap structure in CsFe₂As₂ and KFe₂As₂.

II. EXPERIMENT

The CsFe₂As₂ single crystals were grown by self-flux method for the first time.³⁸ The sample was cleaved to a rectangular shape of dimensions 3.5×1.0 mm² in the ab -plane, with 30 μ m thickness along the c axis. Contacts were made directly on the sample surfaces with silver paint, which were used for both resistivity and thermal conductivity measurements. To avoid degradation, the sample was exposed in air less than 2 hours. The contacts are metallic with typical resistance 100 m Ω at 2 K. In-plane thermal conductivity was measured in a dilution refrigerator, using a standard four-wire steady-state method with two RuO₂ chip thermometers, calibrated *in situ* against a reference RuO₂ thermometer. Magnetic fields were applied along the c axis and perpendicular to the heat current. To ensure a homogeneous field distribution in the sample, all fields were applied at temperature above T_c .

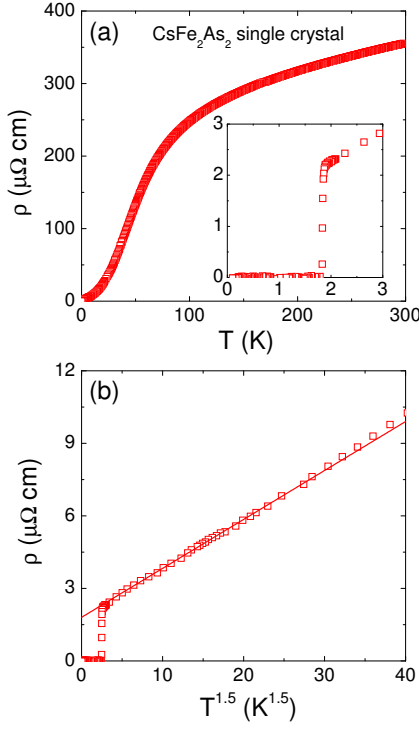


FIG. 1: (Color online). (a) In-plane resistivity of CsFe₂As₂ single crystal. The inset shows the resistive superconducting transition, with $T_c = 1.81$ K defined by $\rho = 0$. (b) Low-temperature resistivity plotted as ρ vs $T^{1.5}$. The solid line is a fit of the data between 2.6 and 9 K to $\rho = \rho_0 + AT^{1.5}$, which gives residual resistivity $\rho_0 = 1.80 \mu\Omega \text{ cm}$.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the in-plane resistivity $\rho(T)$ of CsFe₂As₂ single crystal. The shape of $\rho(T)$ curve mimics that of KFe₂As₂ single crystal.^{29,31} From the inset of Fig. 1(a), the T_c defined by $\rho = 0$ is 1.81 K. For the CsFe₂As₂ polycrystal in Ref. 34, the T_c defined by the sharp drop of susceptibility is about 2.2 K.³⁴ The origin of the 0.4 K difference between the T_c of CsFe₂As₂ single crystal and polycrystal is not clear. One possible reason is that the polycrystal in Ref. 34 is purer than our single crystal, since it has been shown that, for KFe₂As₂, the purer sample has higher T_c .³¹ In any case, the T_c of (K, Rb, Cs)Fe₂As₂ series (3.8 K, 2.6 K, and 1.8-2.2 K, respectively) seems to decrease with the increase of the ionic radius of alkali metal.

In Fig. 1(b), the low-temperature resistivity is plotted as ρ vs $T^{1.5}$. It is found that ρ obeys $T^{1.5}$ dependence nicely above T_c , up to about 9 K. The fit of the data between 2.6 and 9 K gives residual resistivity $\rho_0 = 1.80 \mu\Omega \text{ cm}$, thus the residual resistivity ratio (RRR) = $\rho(300 \text{ K})/\rho_0 \approx 200$ is obtained. For the dirty KFe₂As₂ single crystal with $\rho_0 = 3.32 \mu\Omega \text{ cm}$ and $\text{RRR} \approx 110$, $\rho \sim T^{1.5}$ has already been noticed.²⁹ For the clean KFe₂As₂ single crystal with $\rho_0 = 0.21 \mu\Omega \text{ cm}$ and $\text{RRR} \approx 1180$, $\rho \sim T^{1.8}$ was found.³¹ Such a non-Fermi-liquid behavior of $\rho(T)$

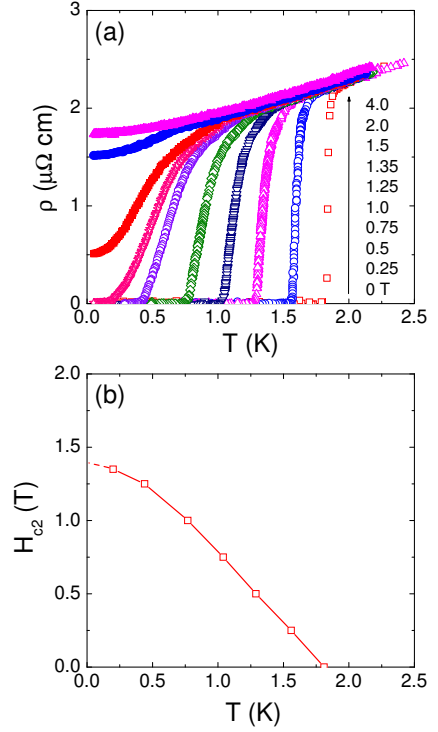


FIG. 2: (Color online). (a) Low-temperature resistivity of CsFe₂As₂ single crystal in magnetic field up to 4 T. (b) Temperature dependence of the upper critical field $H_{c2}(T)$, defined by $\rho = 0$. The dashed line is a guide to the eye, which points to $H_{c2}(0) \approx 1.4$ T.

in KFe₂As₂ and CsFe₂As₂ may result from the antiferromagnetic spin fluctuations.³⁹ In BaFe₂(As_{1-x}P_x)₂, the non-Fermi-liquid linear behavior of $\rho(T)$ near optimal doping, and the increase of power n in the overdoped regime have been considered as the signature of a quantum critical point.⁴⁰

In order to estimate the upper critical field $H_{c2}(0)$ of CsFe₂As₂, the resistivity was also measured in magnetic fields up to $H = 4$ T, as shown in Fig. 2(a). Fig. 2(b) plots the temperature dependence of $H_{c2}(T)$, defined by $\rho = 0$. This definition usually corresponds to the bulk H_{c2} . From Fig. 2(b), we estimate $H_{c2}(0) \approx 1.4$ T. To choose a slightly different H_{c2} does not affect our discussion on the field dependence of κ_0/T below.

The ultra-low-temperature heat transport measurement is a bulk technique to probe the gap structure of superconductors.⁴¹ In Fig. 3(a), we present the temperature dependence of in-plane thermal conductivity for CsFe₂As₂ single crystal in zero and applied magnetic fields, plotted as κ/T vs T . All the curves are roughly linear, as previously observed in dirty KFe₂As₂,²⁹ BaFe_{1.9}Ni_{0.1}As₂,⁹ and Ba(Fe_{1-x}Ru_x)₂As₂ single crystals.²² Therefore we fit all the curves to $\kappa/T = a + bT^{\alpha-1}$ with α fixed to 2. The two terms aT and bT^{α} represent contributions from electrons and phonons, respectively. Here we only focus on the electronic term.

For CsFe₂As₂ in zero field, the fitting gives $\kappa_0/T =$

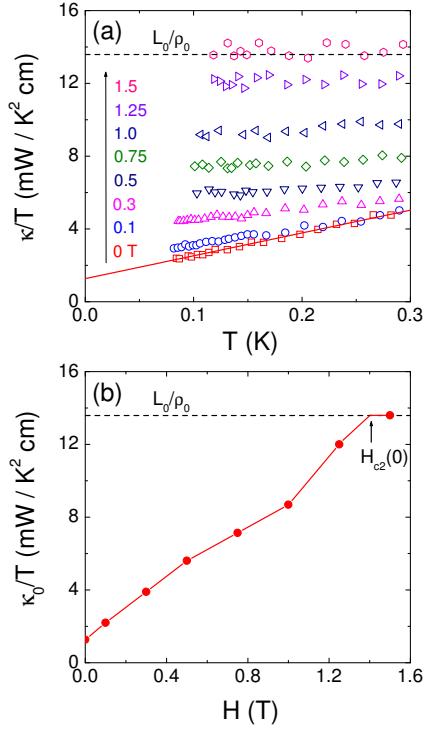


FIG. 3: (Color online). (a) Low-temperature in-plane thermal conductivity of CsFe₂As₂ single crystal in zero and magnetic fields applied along the *c* axis. The solid line is a fit of the zero-field data to $\kappa/T = a + bT$, which gives a residual linear term $\kappa_0/T = 1.27 \text{ mW K}^{-2} \text{ cm}^{-1}$. The dash lines are the normal-state Wiedemann-Franz law expectation L_0/ρ_0 , with L_0 the Lorenz number $2.45 \times 10^{-8} \text{ W } \Omega \text{ K}^{-2}$ and $\rho_0 = 1.80 \mu\Omega \text{ cm}$. (b) Field dependence of κ_0/T . In $H = 1.5 \text{ T}$, slightly above $H_{c2}(0) = 1.4 \text{ T}$, the Wiedemann-Franz law $\kappa_0/T = L_0/\rho_0$ is satisfied.

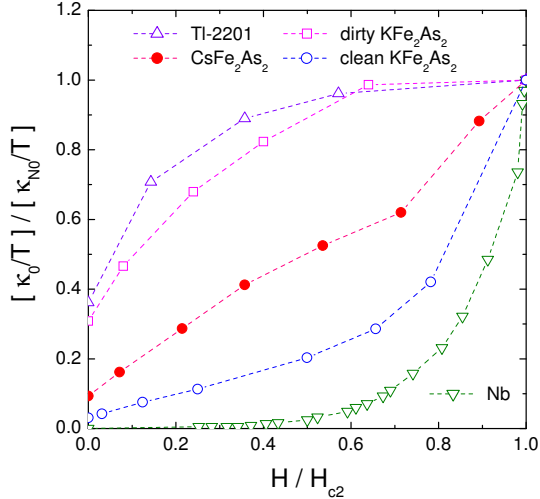


FIG. 4: (Color online). Normalized residual linear term κ_0/T of CsFe₂As₂ as a function of H/H_{c2} . For comparison, similar data are shown for the clean *s*-wave superconductor Nb,⁴⁴ an overdoped *d*-wave cuprate superconductor Tl-2201,⁴² the dirty and clean KFe₂As₂.^{29,31}

$a = 1.27 \pm 0.04 \text{ mW K}^{-2} \text{ cm}^{-1}$. This value is about 1/10 of the normal-state Wiedemann-Franz law expectation $\kappa_{N0}/T = L_0/\rho_0 = 13.6 \text{ mW K}^{-2} \text{ cm}^{-1}$, with L_0 the Lorenz number $2.45 \times 10^{-8} \text{ W } \Omega \text{ K}^{-2}$ and normal-state $\rho_0 = 1.80 \mu\Omega \text{ cm}$. For a high-quality superconductor with no impure phase, such a significant κ_0/T in zero field is usually contributed by nodal quasiparticles, thus considered as a strong evidence for nodes in the superconducting gap.⁴¹ For example, $\kappa_0/T = 1.41 \text{ mW K}^{-2} \text{ cm}^{-1}$ for the overdoped cuprate Tl₂Ba₂CuO_{6+δ} (Tl-2201), a *d*-wave superconductor with $T_c = 15 \text{ K}$.⁴² Previously, $\kappa_0/T = 2.27$ and $3.6 \text{ mW K}^{-2} \text{ cm}^{-1}$ were observed for dirty and clean KFe₂As₂, respectively.^{29,31}

For the clean KFe₂As₂, an additional large electronic term $\kappa/T \sim T^2$ was also observed.³¹ Reid *et al.* considered this term as the leading-order finite-temperature correction to κ/T for *d*-wave superconductor, which will be rapidly suppressed by impurity scattering and magnetic field.³¹ From Fig. 3(a), such an electronic term is absent in our CsFe₂As₂ single crystal, which is not very clean.

The field dependence of κ_0/T can provide further support for the gap nodes.⁴¹ For a nodal superconductor, κ_0/T increases rapidly in low field due to the Volovik effect,⁴³ as in Tl-2201.⁴² In contrast, for a single-gap *s*-wave superconductor, κ_0/T displays a very slow field dependence at low field, as in Nb.⁴⁴ In Fig. 3(b), we plot the field dependence of κ_0/T for CsFe₂As₂. At low field, κ_0/T indeed increases rapidly. Then it shows slight downward curvature before reaching the upper critical field $H_{c2}(0)$. In $H = 1.5 \text{ T}$ slightly above $H_{c2}(0) = 1.4 \text{ T}$, the fitting gives $\kappa_0/T = 13.6 \pm 0.3 \text{ mW K}^{-2} \text{ cm}^{-1}$, satisfying the Wiedemann-Franz law perfectly.

For comparison, the normalized $(\kappa_0/T)/(\kappa_{N0}/T)$ of CsFe₂As₂ is plotted as a function of H/H_{c2} in Fig. 4, together with Nb,⁴⁴ Tl-2201,⁴² the dirty and clean KFe₂As₂.^{29,31} Clearly, the curve of CsFe₂As₂ lies between the dirty and clean KFe₂As₂. The dirty KFe₂As₂ shows similar field dependence of κ_0/T to that of Tl-2201,²⁹ which should also be dirty, with $\rho_0 = 5.6 \mu\Omega \text{ cm}$ and $\text{RRR} \approx 30$.⁴² For the clean KFe₂As₂, Reid *et al.* argued that the field dependence of κ_0/T is a compelling evidence for *d*-wave gap, since the experimental $\kappa_0/T(H)$ curve is close to calculated curve of a *d*-wave superconductor in the clean limit ($\hbar\Gamma/\Delta_0 = 0.1$).³¹ The ρ_0 and RRR of our CsFe₂As₂ lie between the dirty and clean KFe₂As₂, indicating that its impurity level lies between the dirty and clean KFe₂As₂. This may reasonably explain the position and shape of its normalized $\kappa_0/T(H)$ curve in Fig. 4. Such a result suggests that CsFe₂As₂ has a nodal superconducting gap structure similar to that of KFe₂As₂, and shows how the field dependence of κ_0/T evolves with the impurity level.

IV. SUMMARY

In summary, we have measured the thermal conductivity of CsFe_2As_2 single crystal, the sister compound of KFe_2As_2 , down to 50 mK. Both the significant κ_0/T in zero field and the field dependence of κ_0/T provide clear evidence for nodal superconducting gap in CsFe_2As_2 . Our results suggest that the extremely hole-doped (K, Rb, Cs) Fe_2As_2 series of iron-based superconductors should have a common nodal gap structure. More experiments on these compounds are needed to get the consensus on their exact gap symmetry (*d*-wave or

accidental nodal *s*-wave).

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* E-mail: shiyan.li@fudan.edu.cn

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